

URu₂Si₂: hidden order and amplitude of quantum oscillations

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It is shown that the hidden ordered (HO) state in heavy fermionic metal URu₂Si₂ is either nonconventional charge density wave (CDW) or antiferroelectric (AFE) commensurate ordering. Similar to antiferromagnetic (AF) order an antiferroelectric order creates momentum dependence of conducting electrons g -factor. This even modest anisotropy along with the anisotropy of heavy cyclotron mass produce multiple effect of so called zero-spin splitting in the amplitude of the de Haas - van Alphen and the Shubnikov - de Haas signals as function of direction of magnetic field.

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I. INTRODUCTION

During the past three decades, the puzzle of hidden order (HO) in URu₂Si₂ attracts attention of condensed matter community (for the recent reviews see^{1,2} and references therein). This strange state develops below 17.5K. The corresponding electronic specific heat measurements indicates that below this second-order-type transition about one half of the Fermi surface is removed. Nevertheless the material is still a good metal passing to the superconducting state below 1.4 K.

The de Haas van Alphen oscillations corresponding to three kinds of Fermi surfaces were observed clearly in both the normal and superconducting mixed states of URu₂Si₂³. The frequency of oscillations of so called α band is almost independent from the field direction that corresponds to the practically spherical Fermi surface. At the same time the dHvA amplitude does not change with changes of field direction in the basal tetragonal plane while in the (010) plane, the signal amplitude becomes zero at about 10 different field angles. The authors of Ref.10 relate this observation with phenomenon of so called zero spin-splitting in which the up and down spin contribution to the oscillation cancel out. Namely, the dHvA amplitude is proportional to the spin factor

$$\cos(\pi g m_c^*/2m) \quad (1)$$

depending of g -factor value and cyclotron effective mass m_c^* .^{4,5} The latter measured from the Dingle plot is about 10 times larger than the bare electron mass m . It means that g -factor undergoes noticeable variation as function of the field direction changed from parallel to tetragonal axis to parallel to a -axis.

We note, however, that the effect of multiple nullification of the dHvA signal amplitude as function of magnetic field direction is determined by the product of g -factor and the cyclotron mass. These are quantities averaged over the line limiting the extremal cross section of the Fermi surface. So, in heavy fermionic materials when $m^* \gg m$, even not so strong anisotropy in each of these quantities can produce multiple nullification in the dHvA amplitude as function of direction of magnetic field.

This phenomenon quite recently has been studied by measurements of Shubnikov-de Haas oscillations in the same material.⁶ In addition to previous observations³ there was revealed the similar angular dependence of amplitude of β band. The measured¹⁶ cyclotron masses for magnetic field parallel to c -axis are: $m_\alpha^* = 12.4m$ and $m_\beta^* = 23.8m$. They are proved roughly twice smaller for field parallel to a axis. So, if the g -factor magnitude undergoes similar double or so decrease, the multiple nullification of dHvA signal amplitude is quite understandable.

The g -factor anisotropy arises in a multiband metal possessing centre symmetry due to interband spin-orbital coupling. In case of tetragonal URu₂Si₂ the g -factor presents an uniaxial tensor

$$g_{\alpha\beta}(\mathbf{k}) = g_\perp(\mathbf{k})(\hat{x}_\alpha\hat{x}_\beta + \hat{y}_\alpha\hat{y}_\beta) + g_\parallel(\mathbf{k})\hat{z}_\alpha\hat{z}_\beta. \quad (2)$$

So, the modest scale of g -factor anisotropy can be expected due to spin-orbital coupling without attracting of exotic mechanisms in an attempt to explain seeming Ising anisotropy of g -factors of conducting electrons.

Effective g -factor value determining amplitude of de Haas-van Alphen signal from the particular extremal cross section of the Fermi surface is given by the average the momentum dependent g -factor over the curve encircling this area

$$g_{eff} = \frac{\oint \frac{dl}{v_F(\mathbf{k})} g(\mathbf{k})}{\oint \frac{dl}{v_F(\mathbf{k})}}. \quad (3)$$

The anisotropy of g_{eff} has no direct relationship with the g -factor anisotropy determining so called paramagnetic limiting field for superconducting state as it was proposed in recent publications⁷. The latter is determined by g -factors extracted from the magnetic susceptibility proportional to the average of the product of square of local $g(\mathbf{k})$ factor and the local density of states $N_0(\mathbf{k})$ over the whole Fermi surface area including all band sheets. The whole susceptibility can also include the van Vleck component. So, the anisotropy in " g -factor" extracted from susceptibility has less common with anisotropy of de Haas-van Alphen g -factor. It is also pertinent to mention here, that although the susceptibility in URu₂Si₂ along c -axis is three times larger than along a -axis⁸ it

is not an argument to think that the suppression of superconducting state in URu₂Si₂ is determined by pure paramagnetic mechanism.

An attempt to explain the g -factor anisotropy has been undertaken by virtue of arising of "hastatic" ordering that breaks double time-reversal symmetry, mixing states of integer and half-spin.⁹

In contrast to this exotic approach recently on the basis of relativistic density functional theory there was shown that the bandlike $5f$ electrons in URu₂Si₂ exhibit colossal Ising behavior.¹⁰ The origin of the peculiar anisotropy is found due to specific Fermi surface structure and the strong spin-orbital interaction. The calculations¹⁰ has been performed for the AF phase where according to Ref.16 the Fermi surface is practically identical to that of the HO phase.

We have already argued that multiple nullification of dHvA amplitude in a heavy fermionic metal can be explained taking into account some modest anisotropy of effective g -factor arising due to interband spin-orbital interaction. Here we will show that the g -factor anisotropy inevitably appears even in a single band metal due to formation of specific commensurate charge density wave. In the next section it is shown that is the most plausible candidate for the URu₂Si₂ hidden ordered state is commensurate antiferroelectric ordering. The source of g -factor anisotropy based on argumentation has been proposed several years ago by R.Ramazashvili^{11,12} that a commensurate antiferromagnetic order leads to momentum dependence of conducting electrons g -factor. One can come to similar conclusions for a metal with antiferroelectric ordering as well. This will be done in the third section followed by conclusion.

II. HIDDEN ORDER

There was proposed a lot of theories to explain the HO phenomenon (see reviews^{1,2}). Meanwhile several recent and not so recent experimental developments are able to put serious restrictions on theoretical phantasies. In fact the symmetry of HO state is practically fixed by experimental observations.

URu₂Si₂ is a tetragonal material with uranium atoms forming body centered tetragonal lattice. There was found¹³ that at pressure 0.5 GPa at low enough temperatures HO state abruptly transforms to two-sub-lattice antiferromagnetic state. The magnetic moments of the order $0.4 \mu_B$ aligned parallel and antiparallel to c -axis are disposed on uranium atoms with ordering vector $\mathbf{q} = (1, 0, 0)$ such that the body-centered tetragonal structure transforms to the simple tetragonal one. Further investigations demonstrated that at all temperatures HO and AF state are separated from each other by the first order type transition, that is the first order transition line is finished at some bicritical point on the line of the second-order-type transition $T_c(P)$.¹⁴ This observation has the important consequence: the HO state and

AF state have to have the different symmetries. This is because at their symmetry coincidence, the line of the first order transition should obligatory terminate at some critical point below $T_c(P)$ as it was demonstrated in the paper¹⁵.

Another important observation relating to symmetry of HO state was done by measurements of Shubnikov-de-Haas effect on high quality URu₂Si₂ single crystals.¹⁶ Namely, there was shown that under pressure for field $H \parallel \hat{z}$ the Fermi surface reveals only minor changes between the HO state and AF state. This was strong indication that both phases have the same unit cell doubling and the same ordering vector.

The change in the electronic periodicity at the transition from the normal body-centered tetragonal state to the simple tetragonal HO state has been revealed recently by ARPES¹⁷⁻¹⁹ and polarization resolved Raman spectroscopy^{20,21} measurements. Thus, the similarity between HO phase and the high pressure AF phase found in quantum-oscillation experiments has been confirmed.

So, we come to the conclusion that normal and HO states have different translational symmetries and the HO and AF states have the same translational and rotational symmetry but at the same time these phases should be symmetrically different. This case, there is only one opportunity for hidden ordering. It is non-conventional commensurate charge density wave (CDW) along vector $\mathbf{q} = (1, 0, 0)$, that is periodic in space ordering of multipole charge distributions around the uranium sites possessing of local tetragonal symmetry. The appearance of such type ordering has to change magnetization distribution induced in a single crystal of URu₂Si₂ under a magnetic field applied along the tetragonal c axis that has been observed and reported in Ref.22.

The nonconventional commensurate spin density wave (SDW) as candidate for the HO is forbidden because this case the HO and AF phases will have the same symmetry. On the contrary the symmetry allows nonconventional CDW ordering including local breaking of the space parity like a periodic distribution of multipoles alternating by their mirror reflections. This case the initial normal state body centered tetragonal lattice transforms below 17.5 K to simple tetragonal material formed by two sub-lattices differing from each other by the space inversion. Such type commensurate chiral density wave ground state has actually been proposed in Ref. 20. We shall call this state antiferroelectric state (AFE). This state can in general include also the usual antiferromagnetic component such that two sub-lattices transfer each other by application both space and time inversion and shift on vector $\mathbf{q} = (1, 0, 0)$.

Thus, the HO state is either nonconventional CDW or AFE commensurate ordering. In the former case we deal with the simple doubling of body-centered tetragonal unit cell modulated by charge distribution, in the latter, the body-centered lattice consists of two sub-lattices differ each other at least by the space parity transformation. We shall demonstrate that this gives rise to mo-

mentum dependence of electron g -factor revealing itself in the dependence of the amplitude of quantum magnetic oscillations from the direction of magnetic field.

III. G-FACTOR ANISOTROPY IN ANTIFERROELECTRIC STATE

The modification of electron spectrum in single band metal caused by an antiferroelectric (AFE) ordering doubling the initial period of crystal lattice can be derived introducing the Rashba-Bychkov modulation in one-electron Hamiltonian

$$H_{AFE} = \sum_{\mathbf{k}} \left[(\varepsilon_{\mathbf{k}} \delta_{\alpha\beta} - \mathbf{h} \boldsymbol{\sigma}_{\alpha\beta}) a_{\mathbf{k}\alpha}^{\dagger} a_{\mathbf{k}\beta} + i \mathbf{l}_{\mathbf{k}} \boldsymbol{\sigma}_{\alpha\beta} (a_{\mathbf{k}+\mathbf{q}/2,\alpha}^{\dagger} a_{\mathbf{k}-\mathbf{q}/2,\beta} - a_{\mathbf{k}-\mathbf{q}/2,\alpha}^{\dagger} a_{\mathbf{k}+\mathbf{q}/2,\beta}) \right]. \quad (4)$$

Here,

$$\mathbf{h} = g\mu_B \mathbf{H}/2 \quad (5)$$

is constant magnetic field acting on electron spins, $\boldsymbol{\sigma}$ are the Pauli matrices. For simplicity we keep only one harmonic in AFE modulation. Its amplitude is determined by real pseudovector $\mathbf{l}_{\mathbf{k}}$ which satisfies $\mathbf{l}_{\mathbf{k}} = -\mathbf{l}_{-\mathbf{k}}$ and $g\mathbf{l}_{g^{-1}\mathbf{k}} = \mathbf{l}_{\mathbf{k}}$ where g is any symmetry operation in the point group D_4 not including space inversion.

It is instructive to compare the hamiltonian (4) with corresponding hamiltonian of a single-band metal with single harmonic antiferromagnetic (AF) modulation

$$H_{AF} = \sum_{\mathbf{k}} \left[(\varepsilon_{\mathbf{k}} \delta_{\alpha\beta} - \mathbf{h} \boldsymbol{\sigma}_{\alpha\beta}) a_{\mathbf{k}\alpha}^{\dagger} a_{\mathbf{k}\beta} + \boldsymbol{\sigma}_{\alpha\beta} (\mathbf{h}_{\mathbf{k}}^s a_{\mathbf{k}+\mathbf{q}/2,\alpha}^{\dagger} a_{\mathbf{k}-\mathbf{q}/2,\beta} + \mathbf{h}_{\mathbf{k}}^{s*} a_{\mathbf{k}-\mathbf{q}/2,\alpha}^{\dagger} a_{\mathbf{k}+\mathbf{q}/2,\beta}) \right]. \quad (6)$$

Here, the amplitude of the staggered field $\mathbf{h}_{\mathbf{k}}^s = \mathbf{h}_{-\mathbf{k}}^s$ is an even function of the momentum.

The difference of two hamiltonians is the following : the AFE hamiltonian is time reversal symmetric but it breaks the space inversion symmetry, on the contrary, the AF hamiltonian breaks the time inversion but it keeps the space parity.²³ These difference, however, is not important for electron energy bands dispersion: in both cases it is described by equivalent equations.

To find the band energies for AFE one must diagonalize the energy matrix

$$\hat{E} = \begin{pmatrix} \varepsilon_{\mathbf{k}+\mathbf{q}/2} - \mathbf{h} \boldsymbol{\sigma} & i \mathbf{l}_{\mathbf{k}} \boldsymbol{\sigma} \\ -i \mathbf{l}_{\mathbf{k}} \boldsymbol{\sigma} & \varepsilon_{\mathbf{k}-\mathbf{q}/2} - \mathbf{h} \boldsymbol{\sigma} \end{pmatrix}. \quad (7)$$

For simplicity we write the corresponding band energies for two particular field directions along and perpendicular tetragonal axis.

So, for $\mathbf{h} \parallel \hat{z}$ we obtain

$$\mathcal{E}_{h_z} = \varepsilon_{\pm} \pm \sqrt{l_x^2 + l_y^2 + \left(\sqrt{\varepsilon_{\pm}^2 + \mathbf{l}_z^2} \pm h_z \right)^2}, \quad (8)$$

and for $\mathbf{h} \parallel \hat{x}$

$$\mathcal{E}_{h_x} = \varepsilon_{\pm} \pm \sqrt{l_y^2 + l_z^2 + \left(\sqrt{\varepsilon_{\pm}^2 + \mathbf{l}_x^2} \pm h_x \right)^2}. \quad (9)$$

Here

$$\varepsilon_{\pm} = \frac{\varepsilon_{\mathbf{k}+\mathbf{q}/2} \pm \varepsilon_{\mathbf{k}-\mathbf{q}/2}}{2}. \quad (10)$$

Thus, the initial band in field absence splits on two bands due to AFE period doubling. Each of these bands splits on two under magnetic field.

Let us assume that basal plane spin orbital coupling $\mathbf{l}_{\perp}^2 = l_x^2 + l_y^2$ is negligibly small. Then for $\mathbf{h} \parallel \hat{z}$

$$\mathcal{E}_{h_z} = \varepsilon_{\pm} \pm \left(\sqrt{\varepsilon_{\pm}^2 + \mathbf{l}_z^2} \pm h_z \right), \quad (11)$$

and we see, that magnetic field band splitting does not undergo a change, that means g -factor is completely the same as in absence of spin-orbital interaction:

$$g_{\parallel} = 2. \quad (12)$$

On the contrary for field $\mathbf{h} \parallel \hat{x}$ we have in linear in field approximation

$$\mathcal{E}_{h_x} \approx \varepsilon_{\pm} \pm \left(\sqrt{\varepsilon_{\pm}^2 + \mathbf{l}_z^2} \pm \frac{|\varepsilon_{\pm}|}{\sqrt{\varepsilon_{\pm}^2 + \mathbf{l}_z^2}} h_x \right). \quad (13)$$

If the magnitude $|\varepsilon_{\pm}|$ on the corresponding de Haas - van Alphen orbit is much smaller than the spin-orbit amplitude $|\mathbf{l}_z|$ the spin splitting proves to be negligibly small:

$$g_{\perp} \ll g_{\parallel}. \quad (14)$$

So, by comparison of equations (11) and (13) we come to the conclusion of g -factor anisotropy.

IV. CONCLUSION

The whole body of experimental developments including the de Haas - van Alphen and the Shubnikov- de Haas measurements, ARPES and Raman spectroscopy and polarized neutron scattering allows to fix the order parameter of so called hidden order in URu₂Si₂ as nonconventional charge density wave or antiferroelectric commensurate ordering. It is shown that like the antiferromagnetism the antiferroelectric modulation causes essential quasi-momentum dependence of gyromagnetic factor of conducting electrons. Depending on particular band structure and spin-orbital interaction the g -factor anisotropy can be strong or weak. However, in heavy fermionic metals even modest anisotropy of g -factor produce effect of zero-spin splitting manifesting itself in multiple nullification in the amplitude of the de Haas - van Alphen and the Shubnikov - de Haas signals as function of direction of magnetic field.

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